Infrared polaritonics at low temperatures – pushing the limits of light matter coupling.

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The lifetime in surface polaritons presents a fundamental limit for the strength of light matter interactions achievable. This is inherently tied to the decay mechanisms for the polaritons in the material – electron-electron scattering for metals, or acoustic phonon scattering for optical phonons. Many of the decay pathways for polaritons can be managed through reductions in operational temperature, and new states of matter can be accessed. For example, ballistic plasmon polaritons in monolayer graphene can be accessed at cryogenic temperature [1], and structural phase transitions such as charge density waves in transition metal dichalcogenides [2]. For traditional applications of surface polaritons, including sensor and catalytic technologies, the cost of such cryocooling renders these phenomena purely of academic interest. However, emerging applications for polaritonics in mid to long wave (2-20µm) infrared optoelectronics [3] and quantum technologies [4], have significantly reduced restrictions on the use of cryocooling. This presents new opportunities for polaritonics in enhancing cooled devices. However, measuring and exploiting the properties of polaritons, especially in 2D materials such as graphene, hexagonal boron nitride and transition metal dichalcogenides proves extremely challenging. This is due to their small size (typically on the order of 10-100µm), and the challenges of tightly focusing infrared light for microscopic studies in the infrared. In this work we develop a custom cryo-FTIR microscope and leverage it to understand the properties of polariton supporting materials at low temperatures Fig 1a. Unlike s-SNOM based cryo nearfield microscopy, we are able to probe the properties of materials quickly over an extremely broad spectrum, leveraging closed cycle cooling. We choose to focus on two specific materials which are of interest in the nanophotonics community – hexagonal boron nitride [5], and ReS2e₂. Both types of flakes are mechanically exfoliated onto Au coated substrates, which provides high contrast for absorptive resonances, and exhibit anisotropic optical properties. Notably, for ReSe₂ we can observe the polarized excitons and their broadening as a function of temperature **Fig 1b.** Simultaneously, for hexagonal boron nitride we are able to track both the mid- and long-wave infrared optical phonon resonances and damping for different isotopic enrichments Fig 1c. When combined with optical fitting, we can determine the dramatic reduction for these materials as they are cooled to cryogenic temperatures. We also discuss how knowledge of these constants can be used in the design of cooled, infrared optoelectronic technologies.



Fig. 1 (a) Design of the cryo-FTIR microscope for high throughput, high resolution infrared spectroscopy, (b) low temperature spectra of TaS_2 flakes, (c) spectroscopy of hBN flakes of differing isotopic enrichment.

References

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